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Pondermotive Forces of Electrical Origin in Liquid Dielectrics

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PONDERMOTIVE FORCES OF ELECTRICAL ORIGIN

IN LIQUID DIELECTRICS

By

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IN LIQUID DIELECTRICS

This thesis is approved as a creditable, independent investigation by a candidate for the degree, Master of Science, and acceptable as meeting the thesis requirements for this degree; but without implying that the conclusions reached by the candidate are necessarily the conclusions of the major department.

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N. A. P.

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INTRODUCTION

For years the expression for the force per unit volume which arises in a liquid dielectric as a result of an electric field has been a source of discussion and controversy. Although this volume force, commonly called the ponderomotive force, was discovered long ago, even today one person is likely to disagree with another on its nature. Several different expressions for this volume force occur in the literature and, of these, two seem to be most prominent. These two expressions arise from different hypotheses and, since all attempts to reduce one expression to the other have failed, they are taken to be distinct. Each of the expressions has had many supporters. Consequently, there has been much theoretical work as well as some experimental work concerning the topic of volume forces in a liquid dielectric.

Although the most recent writers apparently have dispelled much of the mystery which has long shrouded this topic, some questions remain unanswered. Moreover, since the history of the topic reveals that some controversies "apparently" have been settled several times, anyone acquainted with the history necessarily is skeptical regarding general claims made by an author that an issue is settled. The topic will remain open for discussion until there is a long period of widespread acceptance of a single hypothesis or point of view.

Therefore, the author of this paper has deemed it a worth-while project to construct, from all the pertinent literature available, a history of the developments and recurring controversies concerning this topic along with a large number of references to original works. An interested person can gain from this history an appreciation of the problems and subtleties encountered in the search for an acceptable point of view. The history also could serve as a starting point for a study of the topic.

If it were possible, it would be desirable to end the controversies which exist between the two expressions for the volume force by devising an experiment which could establish the validity of one expression and disprove the other expression. The possibility of accomplishing this is discussed in another section of this paper entitled THE FEASIBILITY OF A DISCRIMINATING EXPERIMENT. In this section two thought experiments are analytically investigated in detail, other experiments are briefly discussed, and general arguments are made from the results of these experiments. The conclusions reached are in agreement with the most recent views on the topic.

A derivation of the Helmholtz volume force expression can be found in APPENDIX I. The method employed to derive this expression is essentially that which can be found in Classical Electricity and Magnetics by Panofsky and Phillips (22).

A derivation of the Kelvin volume force expression can be found in APPENDIX II. The method employed to derive this expression is essentially that which can be found in Introductory Electrodynamics For Engineers by Bennett and Crothers (2).

HISTORY

The following is a history of the important developments and recurring controversies concerning the volume force expressions of Sir William Thomson (Lord Kelvin) and Hermann von Helmholtz. The history was constructed primarily from the literature cited in this paper, but miscellaneous sources were used also. In most cases the author gathered the information for this history from primary sources. However, when these sources were not available reputable secondary sources were employed. The history is comprehensive but not complete.

The physical concept of polarization in a dielectric seems to have originated with Faraday (6) in 1837. However, the mathematical formulation of this idea was not accomplished until 1845 when Lord Kelvin (11) extended Poisson's theory of magnetic dipoles to the analogous electrostatic case. Kelvin's hypothesis was

> . . . it seems probable that a dielectric, subjected to electrical influence, becomes excited in such a manner that every portion of it, however small, possesses polarity exactly analogous to the magnetic polarity induced in the substance of a piece of soft iron under the influence of a magnet. (12, p. 32)

Because of its origin, this hypothesis is often referred to in the literature as the Poisson-Kelvin hypothesis of polarization or, commonly, the Kelvin hypothesis. It has led to a consistent theory of dielectric polarization and to the prediction of a net volume force with a corresponding pressure for the case of an inhomogeneous electric field or medium.

Maxwell was largely responsible for the early mathematical development of Faraday's invention, lines of induction. In his Treatise on Electricity and Magnetism (17), Maxwell employed Faraday's lines of induction in the mathematical formulation of a theory of electrical stress in media. He pointed out that, under the influence of an electric field, a dielectric existed in a state of stress which, for a perfect dielectric, was maintained as long as the charge distribution was maintained. The mathematical expression for this stress, in terms of the electric field, agreed with the corresponding expression from Kelvin's theory of polarization.

In 1880, Korteweg (13) proposed a new method of predicting an electrostatic volume force in a polarized dielectric. This method was based on the relatively new concept of conservation of energy. In 1881, Helmholtz published "On the Forces Acting in the Interior of Bodies Subjected to Magnetic or Dielectric Polarization" (9) In this paper he showed that it was possible, without making any hypothesis regarding the inner constitution of the matter, to determine the mechanical forces acting in the interior of polarized bodies. The volume force that this method predicts is commonly referred to as the Helmholtz volume force.

The theoretical discussions of the volume force expressions aroused the interest of an experimenter named Quincke (24, 25, 26) who set out to demonstrate the existence of a volume force in a liquid dielectric. In one experiment which he performed he immersed the disks of an absolute electrometer in a liquid dielectric and determined the force acting on one of the disks. He assumed that the volume force gave rise to a hydrostatic pressure which produced a mechanical force on the suspended plate. From his measurements he concluded that the volume force did exist and that, within the accuracy of the experiment, the corresponding pressure could be calculated from the expression given by Maxwell.

Another experiment which Quincke devised was intended to demonstrate the existence of a volume force perpendicular to the lines of electric field. In this experiment he placed two horizontal, closely spaced, parallel plates in a liquid dielectric. One side of a glass manometer tube was attached to a hole in the upper plate at its center and a means of increasing the air pressure on this side was provided. The air pressure was increased until an air bubble was formed between the plates. After the air pressure was noted, an electric field was established between the plates and it was found that the bubble tended to become smaller. Quincke interpreted this as an indication of a volume force transverse to the lines of electric field.

The theory according to Kelvin and the theory according to Helmholtz both predicted a volume force in a liquid dielectric, and Quincke experimentally demonstrated its existence. However, the two volume force expressions differed in form. When no one was able to reduce one expression to the other it was assumed by some that they represented two distinct points of view, only one of which could be correct. The numerous controversial papers which have been published are a result of this assumption.

Pockels (23) published a paper in 1893 in which he supported the Helmholtz energy method of deriving the volume force expression and presented the current views on the method. He also gave a list of references to older literature on the method.

A few years later, J. Larmor (14) published a critique of the Helmholtz method in which he charged that this method was basically unsound. In particular he criticized Helmholtz for improperly performing a volume integration of the energy density. He contended that it was illegitimate to perform this operation using the method of integration by parts. He also pointed out that the expression obtained contradicted the corresponding expression given by Maxwell.

Some experimental work was carried out by Bouchet (3) in 1915. This work consisted of using the volume force as a means of measuring the dielectric constant of a liquid. The liquid was placed in a vessel of ebonite whose base consisted of a large brass disk, and another large disk was placed so that it could be maintained at an adjustable distance above the surface of the liquid. Microscopic observations of the field free surface were made exterior to the apparatus by means of a glass tube passing through the wall of the ebonite vessel and inclined outside. When a field was established between the brass disks a displacement of the meniscus in the glass tube was observed. This displacement was related to the pressure and, through the volume force, to the dielectric constant. The experiment was successful for some liquids and for others it was not.

Little material of controversial nature was published between 1906 and 1916. However, in 1916, apparently after no direct rebuttal to Larmor's criticisms had appeared, G. H. Livens (15) asserted that the Helmholtz method was fundamentally unsound. He essentially reemphasized the criticisms of Larmor and implied that another error existed in the method of Helmholtz. He stated that Helmholtz was unjustified in equating the work done by the forces acting on the material element during its displacement to the change in energy in the element of volume originally occupied by the element before its displacement. This energy, he emphasised, was not the same as the energy of the moving element itself. He also generalized the method of Helmholtz to the case where no linear relationship existed between the electric displacement vector and the electric field vector and showed that this led to absurd results.

Livens again renewed the controversy in his textbook of 1918 (16). He reiterated the criticisms of Larmor and also his own earlier criticism, but this time in more detail. That he was disturbed over the general acceptance of the method of Helmholtz is obvious from this statement concerning his criticism: "This criticism appears however to have been entirely overlooked and Helmholtz's procedure is still tacitly accepted and reproduced by all recent writers on the subject." (16, p. 187)

Bouchet (4) again made a contribution in 1921 by conducting more experimental research. This time, for his apparatus he chose a rectangular trough, two walls of which were conducting plates and two walls of which were glass. A hole in the bottom of the apparatus communicated with a capillary tube which allowed him to observe, in a field free region, the meniscus of the liquid used. When a field was established in the liquid, the meniscus dropped in the capillary tube, thus indicating a pressure change. However, for some liquids there was a change in the level of this meniscus with time and no account for this change could be made. This did not occur when an alternating field was used.

Only a month later, apparently after reviewing the experiment performed by Bouchet, F. Michaud and A. Balloul (20) published a paper criticizing the experimental techniques of Bouchet and describing similar experiments which they had performed. By using a cylindrical condenser with an outside capillary tube they were able to reproduce their results accurately to within 0.2% and did not observe the irregular fluctuation that Bouchet had noted.

A paper was published in 1922 in which the author, G. Gouy (7), showed that the apparent reduction of the real electrostatic forces within a liquid dielectric could be accounted for by considering the action of the pressure which arises from the electric volume force. The old assumption that the real electrostatic forces varied with the dielectric constant thus could be discarded.

In his textbook published in 1941, Stratton (30, p. 146) made the following comments about the criticisms that the Helmholtz method had received:

> These criticisms, however, do not appear to be well founded. Objections to the particular form of the energy integral employed by Helmholtz are satisfied by a more careful procedure. Livens has undertaken a generalization of the energy method to media in which the relation between $\bar{D}$ and $\bar{E}$ is nonlinear in order to show that it leads to absurd results; in so doing he has omitted the essential term associated with the deformation. . . . There appears to be little reason to doubt that the energy method of Korteweg and Helmholtz is fundamentally sound.

In 1949, Smith-White published a paper concerning these electric volume force expressions. In this paper he argued that the Helmholtz method was fundamentally unsound and that the Kelvin hypothesis should be adopted as a starting point for a consistent theory of electrostatics.

The paper charged that in the Helmholtz method the energy associated with a deformation of the dielectric was improperly assumed to be an energy function expressible in terms of the coordinates of the system. In addition, the paper charged that, because of the method employed to derive the Helmholtz volume force expression, this expression could not be considered valid for deformable dielectrics. In support of the Kelvin hypothesis, Smith-White derived from the Kelvin volume force expression an expression which he interpreted as a form of the first law of thermodynamics as it would appear when applied to dielectric phenomena.

Again, differences of opinion arose on this topic when Cade (5) published his views criticizing Smith-White's work. Specifically, he criticized Smith-White for using in his volume force expression the macroscopic field instead of the resultant field, sometimes called the local field or the molecular field. Smith-White (28), however, published a letter concerning this criticism and expressed the opinion that the criticism was due to a misunderstanding of Smith-White's theory.

Experimental advancements were made in 1948 by H. Greinacher (8). In that year he published two papers. The first paper concerned a method, similar to that used by L. Boucuet, of measuring dielectric constants of liquids. Greinacher, however, apparently improved the experimental technique necessary for this method, since he was able to obtain excellent results for many liquids. He also was able to demonstrate the direct proportionality between the square of the field strength and the pressure difference which developed because of the volume force. In his second paper of that year he described how he used this pressure difference as an indicator for an electrostatic voltmeter. Essentially, the voltmeter consisted of a capillary tube placed between two parallel plates. When a field was established between the two plates, the meniscus of the liquid which was used in the voltmeter raised in the capillary tube. By viewing the meniscus in the calibrated capillary tube one could obtain a direct reading of the potential difference applied to the parallel plates.

Smith-White (29) published another paper on the topic in 1952. Some of the many claims and charges which he included in his paper are mentioned here. He particularly emphasised that there existed a general misrepresentation of the physical significance of the Kelvin hypothesis and that there were widespread misconceptions concerning the notion of energy in electrical systems. In support of this statement, he emphasised that the Kelvin volume force expression was a hypothesis and derived from it an expression for the energy in an electrical system which was consistent with the first law of thermodynamics. In addition, he calculated the work done in a deformation of a dielectric and showed that the system was not conservative. Thus, he contended that the Helmholtz method was basically unsound because it assumed a conservative system.

He also devoted some space to criticizing particular authors who had written on this topic. He criticized De Donder for not recognising that one of his own expressions was not a potential energy function. He criticized Stratton and also Abraham and Becker for taking an energy formula to be general when it was derived from a particular case. He criticized Lorentz for not recognizing that his stress tensor should be in symmetric form in order for it to be physically significant. He also pointed out that, for the case where the dielectric constant was not directly proportional to the density, the two distinct volume force expressions predicted different pressures.

In 1953, P. Mazur and I. Prigogine (19) published a paper in which they claimed that there was no real basis for a controversy. They interpreted the Helmholtz energy expression as the thermodynamic free energy of the system and derived from it expressions for the total energy, the entropy, and the Helmholtz volume force. They also derived, this time from Kelvin's volume force expression, expressions for the thermodynamic free energy, the total energy, and the entropy. These expressions were identical to the corresponding expressions used in the Helmholtz method. Regarding this result, the authors made the following statement:

> Le résultat essentiel de ce chapitre est de montrer la complète équivalence des conceptions de Helmholtz et de Kelvin pour la thermodynamique des diélectriques, contrairement à l'opinion exprimée par Smith-White. (19, p. 6)

Although they did not develop this idea in their paper, they implied that the key to the controversial nature of this topic was in the manner in which one separated into its constituent parts the total electromagnetic and pressure tensor associated with the volume force. They also mentioned that the Helmholtz method was valid only for reversible phenomena whereas the Kelvin expression could be applied to irreversible phenomena. No direct answers to Smith-White's criticisms were made nor was any comment made concerning the conditions, if any, under which a liquid dielectric may be considered a reversible system. They did show, however, that Cade's criticism of Smith-White's work was unjustified, since both the macroscopic field and the resultant field yielded the same results.

The most recent paper on the topic seems to be the one published by P. Mazur and S. R. De Groot (18) in 1956 in which they approach the problem from a statistical mechanical point of view. In this paper they developed both the Kelvin and the Helmholtz expressions for the volume force and pointed out that the two cases corresponded to two different ways of defining the pressure and the ponderomotive force. They also pointed out that no matter how one chose to define the pressure the sum of the negative divergence of the pressure tensor and the ponderomotive force must remain invariant. In addition, they commented that the Kelvin expression had a wider range of validity since it was necessary to assume statistical equilibrium in the derivation of the Helmholtz expression.

Although textbooks also are divided on this topic, they usually do not discuss the controversies. Probably this is because space cannot be set aside for a specialized discussion when a large amount of material is to be covered. Some of the textbook authors who have adopted Kelvin's hypothesis are Livens (16), Bennett and Crothers (2), and Page (21). Some authors who seem to prefer the Helmholtz point of view are Jeans (10), Abraham and Becker (1), Stratton (23), and Panofsky and Phillips (22).

THE FEASIBILITY OF A DISCRIMINATING EXPERIMENT

It is logical to suppose that many attempts have been made to devise an experiment that could determine which of the two distinct expressions for the volume force was not in accordance with experimental observations. Obviously none of these attempts have succeeded since such an accomplishment surely would have been conspicuously published. In this section thought experiments with liquid dielectrics are investigated in an effort to discover why such a discriminating experiment has never been devised. In the course of this investigation the details of the spatial variation in the pressure of electrical origin are revealed, pressure measuring schemes are very briefly discussed, and a plausible conclusion is reached regarding the feasibility of a discriminating experiment.

The MKS rationalized system of units is used consistently throughout this paper. All symbols used in this paper are defined in Table I. Derivations of the two volume force expressions are located in the appendices.

Experiment One

Consider an experimental apparatus similar to that employed by L. Bouchet (see the section of the paper entitled HISTORY) in 1921. The apparatus is shown schematically in Figure 1. It will be helpful to identify the several points which have been assigned a letter in Figure 1. The point A has been chosen as a point just above the interface between the liquid dielectric and air. The dielectric constants

Table I

Definition of Symbols

g	acceleration of gravity
ρ'	charge density
K	dielectric constant
$\bar{D}$*	displacement vector
P	electrical pressure of Helmholtz
p'	electrical pressure of Kelvin
$\bar{f}$	force on a dipole
$\bar{\nabla}$	gradient operator
h	height of liquid column
$\bar{E}$	macroscopic electric field vector
ρ	mass density
$\bar{e}$	microscopic electric field vector
$\bar{p}$	molecular dipole moment
ϵ_o	permittivity of free space
$\bar{P}$	polarization vector
F	thermodynamic free energy
$\delta\bar{s}$	virtual displacement
$\bar{u}$	virtual velocity
$\bar{F}$	volume force of Helmholtz
$\bar{F}'$	volume force of Kelvin

*A bar over a symbol denotes a vector quantity.

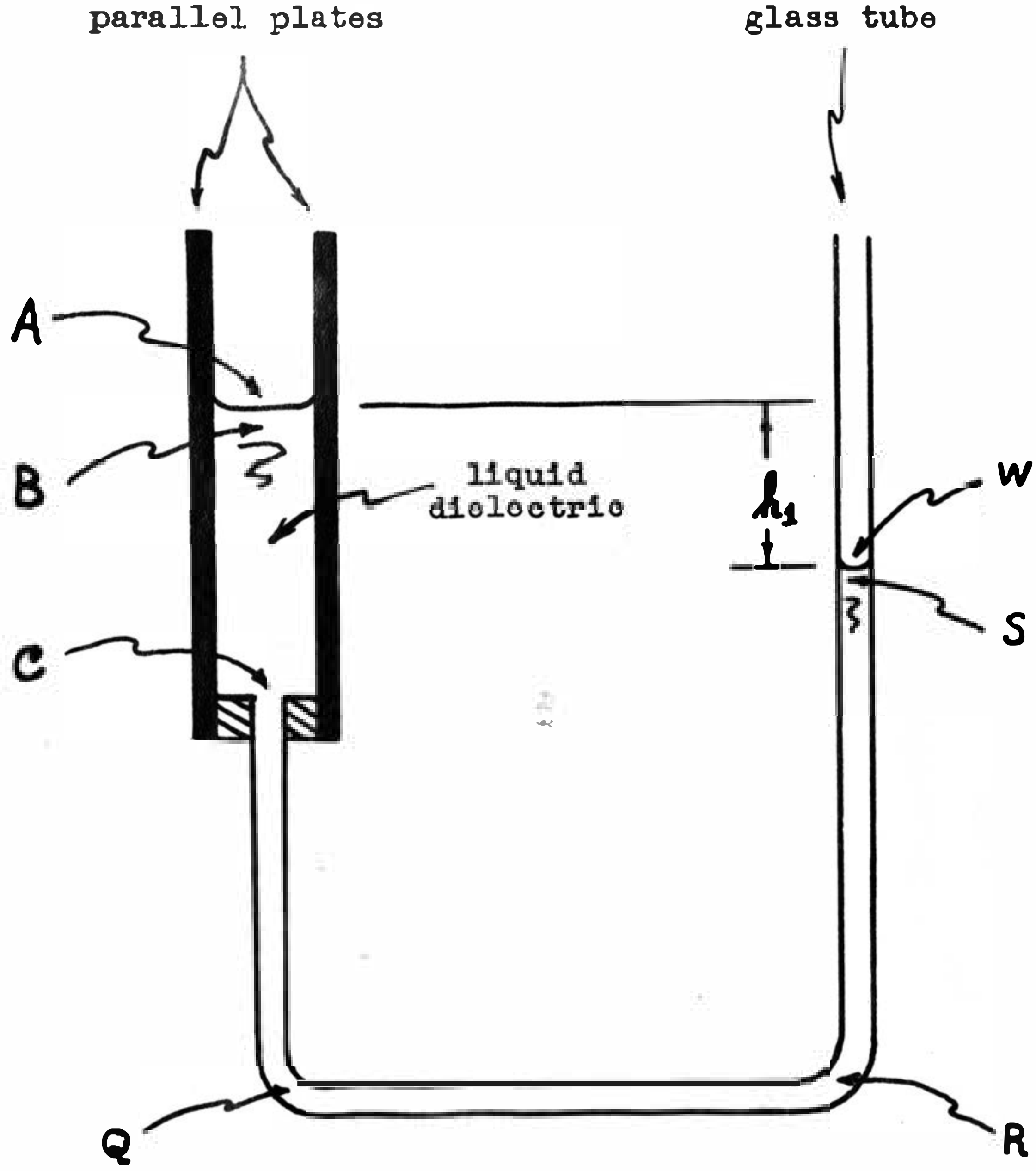

Figure 1. Apparatus for Experiment One

of air and the liquid used are taken to be approximately one and K respectively. Directly below A is a transition region between air and liquid in which the density and dielectric constant change rapidly but continuously with depth. The change can be considered continuous even though the transition region may be as small as one or two molecular diameters. This is so since the molecules of the dielectric will not be arranged in a definite row at the interface. There will exist spaces within the dielectric very near the interface where molecules do not exist, and also there will be molecules projecting above the interface into the air filled region. Therefore a continuous average density is sensible. The point B is identified as a point just below A and just below the transition region. The point C is that point below B which marks the end of the region of homogeneous field and the beginning of the inhomogeneity in the field. Somewhere between C and Q the field becomes homogeneous again and takes on the value zero. The point Q is merely a point which is below C and is in a zero field. The point R is a point laterally displaced from Q, and the point R along with all points on a line joining Q and R is in a field free region. All of the points directly above R are also in a field free region. The point S is identified as a point just below the transition region associated with the air-dielectric interface. Just above S and the transition region is the point W.

Application of the Helmholtz expression. Consider first the application of the Helmholtz expression to this particular case. This expression is stated here as equation (1).

$$\bar{F} = -\frac{\epsilon_0}{2} E^2 \bar{\nabla} k + \frac{\epsilon_0}{2} \bar{\nabla}\left(E^2 \rho \frac{dk}{d\rho}\right) \tag{1}$$

When an electric field is established in the dielectric the above force acts. Since it is known experimentally that the liquid comes to equilibrium, a hydrostatic pressure necessarily develops to maintain equilibrium. This pressure is obviously related to the force $\bar{F}$ by the equation

$$\bar{F} - \bar{\nabla} P = 0 \; . \tag{2}$$

This pressure shall be referred to as the pressure of electrical origin or the electrical pressure in order to distinguish it from the pressure due to the gravitational field. The sum of the electrical pressure and the gravitational pressure shall be called the resultant pressure.

A relationship between k and ρ is required since $\frac{dk}{d\rho}$ must be evaluated. Such a relationship is found in the Clausius-Mossotti equation and it will be used here since it has been successfully applied to many liquids. Because this equation is often derived in textbooks (cf. 22, p. 33) it merely will be stated here.

$$\frac{k-1}{k+2} = (\text{constant})\,\rho \tag{3}$$

Differentiating equation (3) leads to the result

$$\frac{dk}{d\rho} = \frac{(k-1)(k+1)}{3\rho} \; . \tag{4}$$

Equations (1), (2), and (4) may be combined to give

$$\bar{\nabla} P = -\frac{\epsilon_0}{2} E^2 \bar{\nabla} K + \frac{\epsilon_0}{6} \bar{\nabla}\left[(k-1)(k+1) E^2\right] \; . \tag{5}$$

It is desirable to learn how the electrical pressure varies along the line which connects the points A, B, C, Q, R, S, and W. This line will be called the path of integration and arbitrary positions on it will be denoted by the symbol x. The electrical pressure associated with a particular point on this line will be denoted by P_x. If only variations along the path of integration are considered equation (5) may be rewritten as

$$\delta P = -\frac{\epsilon_0}{2} E^2 \delta K + \frac{\epsilon_0}{6} \delta\left[(K-1)(K+1) E^2\right] . \tag{6}$$

Equation (6) may be integrated to give

$$\int_A^x \delta P = P_x - P_A = I_1 + I_2 \tag{7}$$

where

$$I_1 = -\frac{\epsilon_0}{2} \int_A^x E^2 \delta K \tag{8}$$

and

$$I_2 = \frac{\epsilon_0}{6} \int_A^x \delta\left[(K-1)(K+1) E^2\right] . \tag{9}$$

The integral I_1 has the value

$$I_1 = -\frac{\epsilon_0}{2}(K_x - 1) E_B^2 \tag{10}$$

for $A < x < B$ since the boundary condition requires that the tangential component of the electric field vector be continuous across a boundary. The notation "$A < x < B$" denotes that x is located between A and B on the path of integration.

For $B < x < C$ the integral I_1 becomes

$$I_1 = -\frac{\epsilon_0}{2}\int_A^B E^2\,dK - \frac{\epsilon_0}{2}\int_B^x E^2\,dK \tag{11}$$

or

$$I_1 = -\frac{\epsilon_0}{2}(K_B - 1)\,E_B^2 \tag{12}$$

since dK is zero for $B < x < C$.

For $C < x < Q$ the integral I_1 becomes

$$I_1 = -\frac{\epsilon_0}{2}\int_A^C E^2\,dK - \frac{\epsilon_0}{2}\int_C^x E^2\,dK \tag{13}$$

or

$$I_1 = -\frac{\epsilon_0}{2}(K_B - 1)\,E_B^2 \tag{14}$$

since dK is zero for $C < x < Q$.

For $Q < x < W$ the integral I_1 becomes

$$I_1 = -\frac{\epsilon_0}{2}\int_A^Q E^2\,dK - \frac{\epsilon_0}{2}\int_Q^x E^2\,dK \tag{15}$$

or

$$I_1 = -\frac{\epsilon_0}{2}(K_B - 1)\,E_B^2 \tag{16}$$

since the field has a zero value for $Q < x < W$.

The integral I_2 may be evaluated simply as

$$I_2 = \frac{\epsilon_0}{6}(K_x - 1)(K_x + 1)\,E_x^2 \tag{17}$$

since the dielectric constant for air is taken to be approximately one.

The results of the preceeding may be expressed in the following form:

For $A < x < B$ $\quad P_x = P_A + \frac{\epsilon_0}{6}(K_x - 1)^2 E_B^2$ (18)

For $B < x < C$ $\quad P_x = P_A + \frac{\epsilon_0}{6}(K_B - 1)^2 E_B^2$ (19)

For $C < x < Q$ $\quad P_x = P_A + \frac{\epsilon_0}{6}(K_B - 1)\left[(K_B + 2)E_x^2 - 3E_B^2\right]$ (20)

For $Q < x < W$ $\quad P_x = P_A - \frac{\epsilon_0}{2}(K_B - 1)\, E_B^2$ (21)

It will be instructive to plot these results on a graph so that one can see at a glance the details of the spatial variation in electrical pressure. Necessarily, however, the scale of this graph will be grossly distorted since variations will be plotted which take place in a space of molecular dimensions. This plot is made in Figure 2.

Figure 2 shows that the electrical pressure rises sharply in the transition region between A and B even though the field is constant there. This sharp increase in pressure is due to the rapid increase in dielectric constant. Between B and C the electrical pressure is maintained at a fixed value since both the field and dielectric constant do not change in this region. Because the field diminishes to zero somewhere between C and Q there is a corresponding drop in electrical pressure in this region. Between Q and W the field is zero and thus the electrical pressure is constant there.

Also shown in Figure 2 is the spatial variation in gravitational pressure. This pressure is obviously a linear function of the depth. Because the transition regions are of molecular dimensions no appreciable

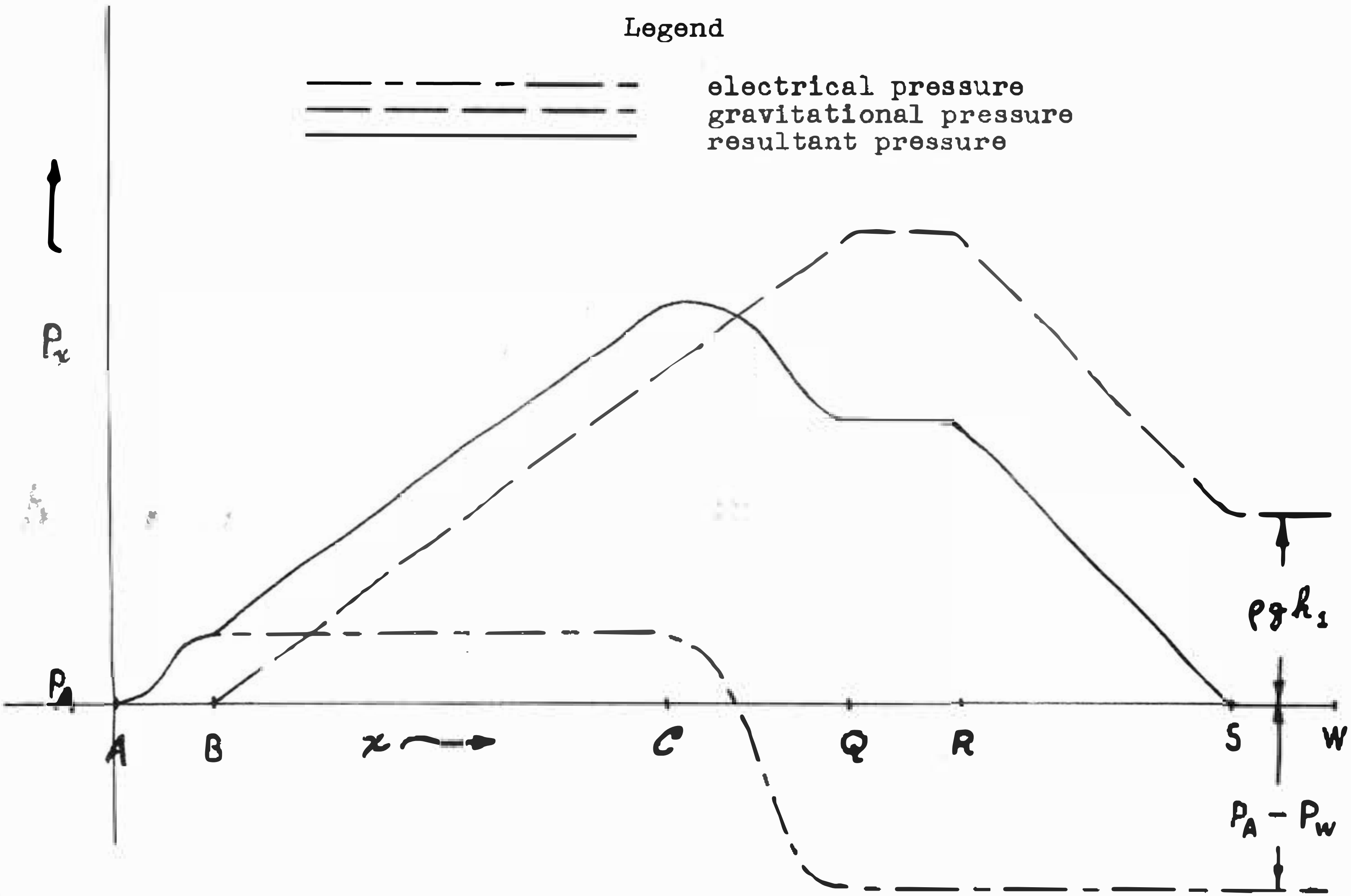

Figure 2. Spatial pressure variations within
the apparatus shown in Figure 1 as predicted by the Helmholtz Method

gravitational pressure change can arise there.

The resultant pressure is obtained by adding the ordinates of the gravitational pressure and the electrical pressure. Since the points A and W are exposed to atmospheric pressure the resultant pressure at these two points must be the same. This requires that the liquid levels adjust until a gravitational pressure difference $\rho g h_1$ is maintained equal to the electrical pressure difference $P_A - P_W$. This may be stated in equation form.

$$\rho g h_1 = P_A - P_W \tag{22}$$

It is clear that the apparatus of experiment one allows one to determine experimentally the electrical pressure difference $P_A - P_W$ and check the Helmholtz volume force expression. This, of course, has been done and the results were in agreement with equation (21).

Application of the Kelvin expression. Consider next the application of the Kelvin expression to this same particular case. The expression is stated here as equation (23).

$$\bar{F}' = \frac{\epsilon_0}{2}(K-1)\,\bar{\nabla} E^2 \tag{23}$$

This volume force likewise produces a hydrostatic pressure. Thus

$$\bar{F}' - \bar{\nabla} P' = 0 \tag{24}$$

or

$$\bar{\nabla} P' = \frac{\epsilon_0}{2}(K-1)\,\bar{\nabla} E^2 . \tag{25}$$

If pressure variations along the path of integration are desired equation (25) may be rewritten as

$$dP' = \frac{\epsilon_0}{2}(K-1)\, d(E^2) \tag{26}$$

and integrated to give

$$\int_A^x dP' = P_x' - P_A' = I \tag{27}$$

where

$$I = \frac{\epsilon_0}{2}\int_A^x (K-1)\, d(E^2) \quad . \tag{28}$$

For $A < x < B$ the integral I becomes

$$I = 0 \tag{29}$$

since $d(E^2)$ does not exist between A and B.

For $B < x < C$ the integral I becomes

$$I = 0 \tag{30}$$

since $d(E^2)$ does not exist between B and C.

For $C < x < Q$ the integral I becomes

$$I = \frac{\epsilon_0}{2}\int_A^C (K-1)\, d(E^2) + \frac{\epsilon_0}{2}\int_C^x (K-1)\, d(E^2) \tag{31}$$

or

$$I = \frac{\epsilon_0}{2}(K_B - 1)\left[E_x^2 - E_B^2\right] \tag{32}$$

since K is constant between C and Q and $\bar{E}_C$ equals $\bar{E}_B$.

For $Q < x < W$ the integral I becomes

$$I = \frac{\epsilon_0}{2}\int_A^Q (K-1)\,d(E^2) + \frac{\epsilon_0}{2}\int_Q^x (K-1)\,d(E^2) \tag{33}$$

or

$$I = -\frac{\epsilon_0}{2}(K_B - 1)\,E_B^2 \tag{34}$$

since E_x^2 is zero in this region.

The results of the preceding may be expressed in the following form:

For $A < x < B$ $\quad P_x' = P_A'$ (35)

For $B < x < C$ $\quad P_x' = P_A'$ (36)

For $C < x < Q$ $\quad P_x' = P_A' + \frac{\epsilon_0}{2}(K_B - 1)\left[E_x^2 - E_B^2\right]$ (37)

For $Q < x < W$ $\quad P_x' = P_A' - \frac{\epsilon_0}{2}(K_B - 1)\,E_B^2$ (38)

These results are shown in Figure 3.

Figure 3 shows that, according to Kelvin's method, the electrical pressure is constant between A and C even though there is a change in dielectric constant in this region. Between C and Q the inhomogeneous field gives rise to a change in electrical pressure, but from Q to W the electrical pressure remains fixed. The spatial variations in the gravitational and resultant pressures are also shown in Figure 3. Since the resultant pressure at the air-liquid interfaces must be atmospheric, the liquid levels must adjust until a gravitational pressure difference $\rho g h_2$ is maintained equal to the electrical pressure difference $P_A' - P_W'$.

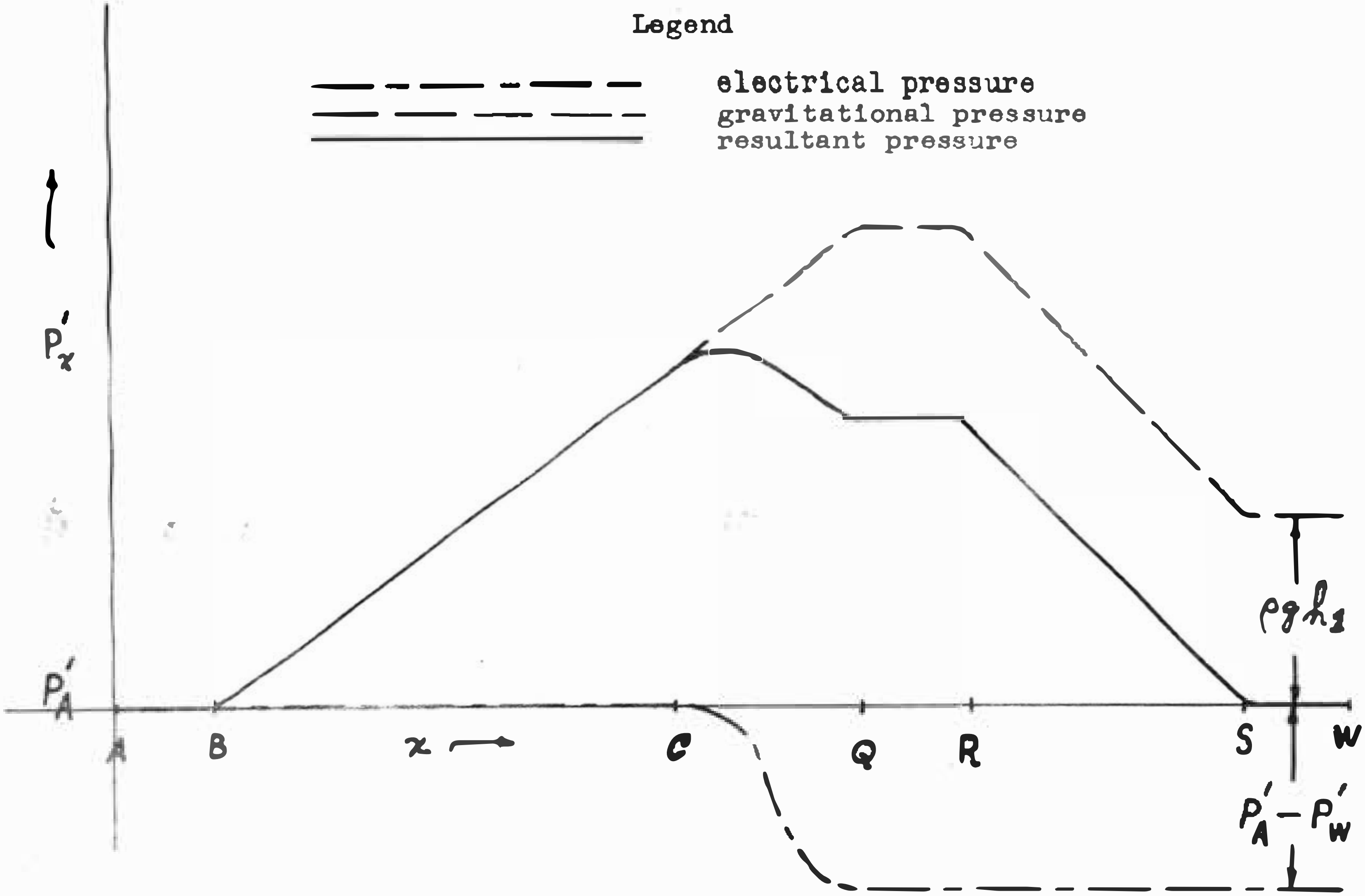

Figure 3. Spatial pressure variations within the apparatus shown in Figure 1 as predicted by the Kelvin Method

Thus, this method too can be checked experimentally by merely measuring the height h_1.

This thought experiment then can be used to check the validity of both volume force expressions for this particular case. The result, however, is that the experiment does not settle any controversy. The Helmholtz method predicts from equation (21)

$$P_A - P_W = \frac{\epsilon_0}{2}(K_B - 1)\, E_B^2 \quad . \tag{39}$$

The corresponding equation (38) of the Kelvin method predicts

$$P_A' - P_W' = \frac{\epsilon_0}{2}(K_B - 1)\, E_B^2 \quad . \tag{40}$$

The right-hand members of equations (39) and (40) are identical. Therefore, for the pressure difference that one is able to measure using this apparatus the two methods predict the same value even though Figures 2 and 3 indicate different pressure variations. This experiment fails as a discriminating experiment.

Experiment Two

For this experiment an apparatus is chosen similar to that employed by L. Bouchet (see the section of this paper entitled HISTORY) in 1915. This apparatus is shown schematically in Figure 4. The letters A, B, S, and W are used to denote points similar to corresponding points in Figure 1.

Since the spatial variation in electrical pressure was investigated in detail for experiment one a detailed investigation will not be presented

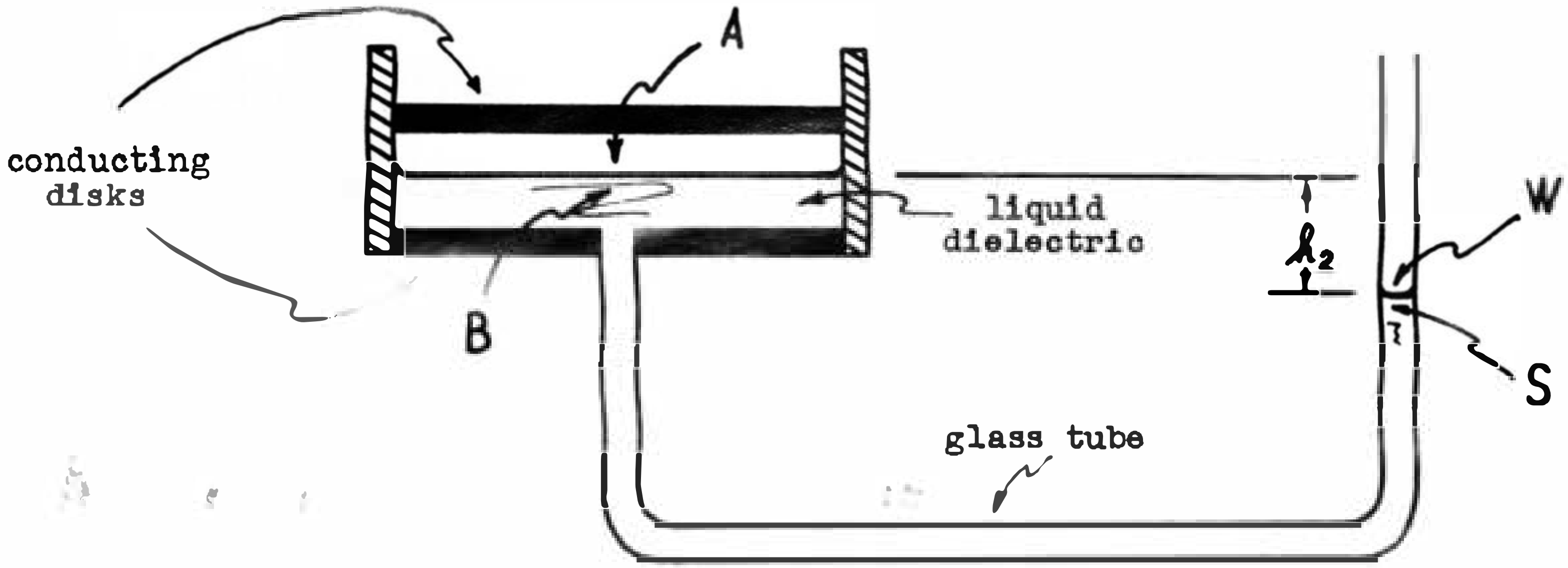

Figure 4. Apparatus for Experiment Two

for experiment two. Such an investigation would reveal a spatial variation in electrical pressure similar to that already discussed.

Application of the Helmholtz expression. Choosing W as the value of χ in equation (8), one can write

$$I_1 = -\frac{\epsilon_0}{2}\left[\int_A^B E^2 dK + \int_B^S E^2 dK + \int_S^W E^2 dK\right] \tag{41}$$

which reduces to

$$I_1 = -\frac{\epsilon_0}{2}\int_A^B E^2 dK \tag{42}$$

since dK does not exist between B and S and E^2 does not exist between S and W. Using the boundary condition that the normal component of the displacement vector must be continuous across a boundary, one can write

$$I_1 = -\frac{\epsilon_0}{2} E_B^2 K_B^2 \int_A^B \frac{dK}{K^2} \tag{43}$$

which can be integrated to give

$$I_1 = -\frac{\epsilon_0}{2} K_B (K_B - 1) E_B^2 \; . \tag{44}$$

Similarly, the integral I_2 may be evaluated as

$$I_2 = 0 \tag{45}$$

since E^2 does not exist at W and K has the value one at A. The desired result is thus

$$P_A - P_W = \frac{\epsilon_0}{2} K_B (K_B - 1) E_B^2 \; . \tag{46}$$

This electrical pressure difference is equal to a gravitational pressure difference $\rho g h_2$ and is subject to experimental measurement since h_2 can be determined.

Application of the Kelvin expression. In applying Kelvin's expression to this case one must evaluate the integral I of equation (28) with x equal to W.

$$I = \frac{\epsilon_0}{2}\left[\int_A^B (K-1)\, d(E^2) + \int_B^S (K-1)\, d(E^2) + \int_S^W (K-1)\, d(E^2)\right] \tag{47}$$

The third term of the right-hand member of this equation vanishes since $d(E^2)$ does not exist between S and W. The second term may be integrated directly to give

$$\frac{\epsilon_0}{2}\int_B^S (K-1)\, d(E^2) = -\frac{\epsilon_0}{2}(K_B-1)\, E_B^2 \; . \tag{48}$$

The first term can be written as

$$\frac{\epsilon_0}{2}\int_A^B (K-1)\, d(E^2) = -\epsilon_0 E_A^2 \int_A^B \frac{(K-1)}{K^3}\, dK \tag{49}$$

after applying the boundary condition

$$E_x = \frac{E_A}{K_x} \; . \tag{50}$$

Equation (49) can be integrated to give

$$\frac{\epsilon_0}{2}\int_A^B (K-1)\, d(E^2) = -\frac{\epsilon_0}{2}\frac{(K_B-1)^2}{K_B^2}E_A^2 = -\frac{\epsilon_0}{2}(K_B-1)^2 E_B^2 \; . \tag{51}$$

The integral I now may be written as

$$I = -\frac{\epsilon_0}{2} K_B (K_B-1)\, E_B^2 \tag{52}$$

and the desired result is

$$P_A' - P_W' = \frac{\epsilon_0}{2} k_B (k_B - 1) E_B^2 \quad . \tag{53}$$

The right-hand members of equations (46) and (53) are identical. Thus for this experiment the two methods predict the same result for the electrical pressure difference which one is able to measure with the apparatus. This experiment, too, fails as a discriminating experiment.

Other Experiments

Of course, any paper must be restricted in its length. For this reason all of the various experiments which have been considered in the preparation of this paper cannot be discussed in detail. However, a few brief comments concerning the other experiments which were considered will be made.

In experiment one the electric field vector was tangent to the liquid-air interface and in experiment two the electric field vector was normal to the liquid-air interface. The more general case in which the electric field vector makes an arbitrary angle with the liquid-air interface was considered. However, no new information was gathered from this spatial arrangement. The electric field vector was resolved into a normal and a tangential component, and the methods used in experiments one and two were applied. The electrical pressure difference determined in this case was equal to the sum of the right-hand members of equations (40) and (46).

In addition to the variation of the spatial relationship between the electric field vector and the liquid-air interface, consideration was given to different schemes for measuring pressure differences. Quincke's schemes (see the section of this paper entitled HISTORY), which involved an absolute electrometer and an air bubble between two parallel plates, were considered and rejected since they failed to discriminate between the two volume force expressions. A U-tube which could be introduced at various locations within a liquid dielectric and which employed either a flexible membrane or an air bubble was considered. This, too, was rejected because it failed to discriminate between the two volume force expressions. A Cartesian diver was also rejected for this reason. In short, the usual schemes of measuring pressure differences seem to fail to discriminate between the two volume force expressions.

CONCLUSIONS

After studying the topic and considering the results of the thought experiments, the author has developed the opinion that any further attempt on his part to devise a discriminating experiment would be fruitless. Moreover, he finds it difficult to imagine that any basically different experiment could be devised to macroscopically distinguish between the two volume force expressions. This, however, is only an opinion based on the following arguments and obviously the results of the preceding section are not conclusive evidence that a discriminating experiment is infeasible.

Consider the Figures 2 and 3 again. These graphs demonstrate the details of the spatial variation in electrical pressure in a liquid dielectric. They also indicate that the two volume force expressions predict different pressures at the same location. In fact they indicate that, if an apparatus could be devised to measure the electrical pressure difference between A and B or the electrical pressure difference between B and W, this apparatus truly could distinguish between the two expressions. However, such an apparatus would have to be especially ingenious, since it would be required to measure a pressure difference which was maintained across a space of molecular dimensions. Moreover, the "usual" pressure measuring devices are inherently doomed to failure since they disturb the very quantity which they attempt to measure. Specifically, they introduce into the region in which one is interested an interface which changes the pressure from what existed before the device was present. This change is just sufficient to allow the results of the application of the two volume force expressions to be identical.

It would seem then that no experimental distinction on a macroscopic scale can be made between the two volume force expressions. In other words, it appears to the author that no real physical distinction exists between the volume forces of Kelvin and Helmholtz, but that the two expressions actually correspond to compatible points of view on the topic of electrical ponderomotive forces in a liquid dielectric.

DISCUSSION

The Figures 2 and 3 show that the electrical pressure associated with the two expressions must be defined differently if one adopts the point of view that the two expressions are compatible. This is apparent also from equations (2) and (24) since the volume force expressions are distinct. Equations (2) and (24) also point out that the sum of the volume force and the negative divergence of the pressure must be invariant. Among other things, this essentially was what Mazur, Prigogine, and De Groot (18, 19) pointed out.

Also, Mazur and De Groot (18) implied that one reason the two expressions differed was because of the existence of short-range and long-range forces between molecules. Possibly this accounts for the difference in pressure variation in the transition region between A and B of Figures 1 and 4. Thus, the results of this paper seem to be in agreement with the most recent papers on the topic.

It is worth-while to comment that further experimental investigation could be carried out on a microscopic scale in the transition region mentioned above. It appears that, if a distinction between the two volume force expressions can be made, it is likely to be made in this region.

SUMMARY

Included in this paper is a comprehensive history of the developments and recurring controversies concerning two distinct expressions for the force per unit volume which arises in a liquid dielectric as a result of an electric field. The two expressions bear the names of the two persons most responsible for their early development, Helmholtz and Kelvin.

The feasibility of devising an experiment which could end the controversies of this topic is discussed. The Helmholtz and the Kelvin volume force expressions are applied to two thought experiments in an effort to discover if these experiments could discriminate between the two expressions. The two experiments fail as discriminating experiments, but in the course of the investigation of the experiments the details of the spatial variations in pressure in the experimental apparatus are revealed. Other possible discriminating experiments are briefly considered but these also fail. The results of these considerations seem to indicate that a discriminating experiment is not feasible.

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APPENDIX I

The volume force expression of Helmholtz is based on the hypothesis that the following volume integral represents the electrical energy stored in a dielectric:

$$F = \frac{1}{2}\int \bar{E} \cdot \bar{D}\, dv \tag{54}$$

It is interpreted as being the thermodynamic free energy of the system.

Since free energy represents the maximum work that can be obtained from a system under isothermal conditions, one may apply the principle of virtual work to equation (54) provided the isothermal condition is met. This means that, if the dielectric in question is surrounded by a heat bath to fix the temperature, the variation in free energy associated with a virtual displacement of a tiny volume dv of dielectric is equal to the negative of the work done by the electrical volume force $\bar{F}$. This can be expressed as

$$\delta F = -\int \bar{F} \cdot \delta\bar{s}\, dv\ ; \tag{55}$$

therefore, the time rate at which free energy is lost from the system is equal to the rate at which work is done.

$$\frac{dF}{dt} = -\int \bar{F} \cdot \bar{u}\, dv \tag{56}$$

Here $\bar{u}$ represents the virtual velocity of the unit volume undergoing the virtual displacement.

Operations will be made on equation (54) until it is in the form of a volume integral of a dot product of the virtual velocity and another vector. This other vector then can be identified as the electrical volume force since the virtual displacement and consequently the virtual velocity are completely arbitrary.

Consider the variation

$$\delta F = \frac{1}{2}\,\delta\int \bar{E}\cdot\bar{D}\,dv \quad . \tag{57}$$

If $\bar{D} = \epsilon_0 k \bar{E}$, then

$$\delta F = \frac{1}{2\epsilon_0}\int \delta\left(\frac{D^2}{K}\right) dv \tag{58}$$

or

$$\delta F = \frac{1}{2\epsilon_0}\left[\int D^2\,\delta\left(\frac{1}{K}\right) dv + \int \frac{1}{K}\,\delta D^2\,dv\right] \quad . \tag{59}$$

If the discussion is restricted to the case where ρ' is zero and no variation in ρ' is allowed, then

$$\delta D^2 = 0 \tag{60}$$

since

$$\delta\rho' = \delta\bar{\nabla}\cdot\bar{D} = \bar{\nabla}\cdot\delta\bar{D} \tag{61}$$

Equation (59) now may be written as

$$\delta F = \frac{1}{2\epsilon_0}\int D^2\left(-\frac{1}{K^2}\right)\delta k\,dv = -\frac{\epsilon_0}{2}\int E^2\,\delta k\,dv \tag{62}$$

or

$$\frac{dF}{dt} = -\frac{\epsilon_0}{2}\int E^2\,\frac{\partial k}{\partial t}\,dv \quad . \tag{63}$$

It should be pointed out that $\frac{\partial K}{\partial t}$ represents the time rate of change of dielectric constant due only to the change in state of the dielectric associated with the virtual displacement.

It will be assumed that the dielectric constant can be expressed as a function of the density only.

$$K = \phi(\rho) \tag{64}$$

The density, however, is a function of other variables. Ordinarily the density would be a function only of the coordinates, say x, y, and z. However, if one considers a virtual displacement and a corresponding virtual velocity the density is not only a function of the coordinates but also a function of time explicitly and implicitly. This is so since the deformation of the dielectric changes the density of the volume element considered and in addition, the coordinates of this volume element change with time. This may be stated mathematically.

$$\rho = \rho(x, y, z, t) \tag{65}$$

with

$$x = x(t, u_x)\,; \quad y = y(t, u_y)\,; \quad z = z(t, u_z)\,. \qquad \text{(66 a,b,c)}$$

Similarly

$$K = K(x, y, z, t)\,. \tag{67}$$

It is required to find how K and thus ρ varies with time explicitly. When such functionality exists as defined by equations (65), (66), and (67), the time rate of change of ρ is called the substantial derivative. It shall be given the symbol $\frac{D}{\partial t}$.

$$\frac{D\rho}{\partial t} = \frac{\partial \rho}{\partial x}\frac{\partial x}{\partial t} + \frac{\partial \rho}{\partial y}\frac{\partial y}{\partial t} + \frac{\partial \rho}{\partial z}\frac{\partial z}{\partial t} + \frac{\partial \rho}{\partial t} \tag{68}$$

or

$$\frac{D\rho}{\partial t} = \bar{\nabla}\rho \cdot \bar{u} + \frac{\partial \rho}{\partial t} \, . \tag{69}$$

Similarly

$$\frac{DK}{\partial t} = \bar{\nabla}K \cdot \bar{u} + \frac{\partial K}{\partial t} \, . \tag{70}$$

It is desirable to express the second term of the right-hand member of equation (70) in terms of the density. The relationship

$$\frac{DK}{\partial t} = \frac{dK}{d\rho}\frac{D\rho}{\partial t} \tag{71}$$

may be used with equation (70) to give

$$\frac{\partial K}{\partial t} = \frac{dK}{d\rho}\left[\bar{\nabla}\rho \cdot \bar{u} + \frac{\partial \rho}{\partial t}\right] - \bar{\nabla}K \cdot \bar{u} \, . \tag{72}$$

Conservation of mass requires that

$$\frac{\partial \rho}{\partial t} = -\bar{\nabla} \cdot (\rho \bar{u}) \tag{73}$$

which leads to

$$\frac{\partial K}{\partial t} = \frac{dK}{d\rho}\left[\bar{\nabla}\rho \cdot \bar{u} - \bar{\nabla} \cdot \rho \bar{u}\right] - \bar{\nabla}K \cdot \bar{u} \, . \tag{74}$$

The identity

$$\bar{\nabla} \cdot (\rho \bar{u}) = \rho \bar{\nabla} \cdot \bar{u} + \bar{u} \cdot \bar{\nabla}\rho \tag{75}$$

may be employed to give

$$\frac{\partial k}{\partial t} = -\frac{dk}{d\rho}\rho\bar{\nabla}\cdot\bar{u} - \bar{\nabla}K\cdot\bar{u} \quad . \tag{76}$$

Equation (63) now may be written in the form

$$\frac{dF}{dt} = \frac{\epsilon_0}{2}\int E^2\frac{dk}{d\rho}\rho\bar{\nabla}\cdot\bar{u}\,dv + \frac{\epsilon_0}{2}\int E^2\,\bar{\nabla}k\cdot\bar{u}\,dv \quad . \tag{77}$$

One may employ an identity similar in form to equation (75) to write

$$\int E^2\frac{dk}{d\rho}\rho\bar{\nabla}\cdot\bar{u}\,dv = \int\bar{\nabla}\cdot\left(E^2\frac{dk}{d\rho}\rho\bar{u}\right)dv - \int\bar{\nabla}\left(E^2\frac{dk}{d\rho}\rho\right)\cdot\bar{u}\,dv \quad . \tag{78}$$

The first right-hand member of equation (78) may be transformed to a surface integral and written as

$$\int_A E^2\frac{dk}{d\rho}\rho\bar{u}\cdot d\bar{A} = 0 \tag{79}$$

since E^2 diminishes as the inverse fourth power and A increases as the square power of the distance from the charges producing $\bar{E}$ to the element of area $d\bar{A}$. Finally, equation (77) may be written as

$$\frac{dF}{dt} = \frac{\epsilon_0}{2}\int\left[E^2\,\bar{\nabla}k - \bar{\nabla}\left(E^2\frac{dk}{d\rho}\rho\right)\right]\cdot\bar{u}\,dv \quad . \tag{80}$$

From equation (56) this identifies $\bar{F}$ with

$$\bar{F} = -\frac{\epsilon_0}{2}E^2\,\bar{\nabla}K + \frac{\epsilon_0}{2}\nabla\left(E^2\frac{dk}{d\rho}\rho\right) \quad . \tag{81}$$

APPENDIX II

The volume force expression of Lord Kelvin is based on the hypothesis that, under the action of an outside charge distribution, the molecules of a dielectric become polarized. It is assumed also that, if the molecules already possess a permanent dipole moment, the imposed electric field creates an average allignment of these dipoles in the direction of the field.

If the electric field is macroscopically inhomogeneous or if the medium itself is inhomogeneous there will be a net force acting on each dipole where this inhomogeneity exists. The expression for this force can be derived from the following arguments.

Consider a region in a dielectric medium which contains many molecules of the dielectric and which is large enough so that a statistical average of the space variations and the time variations of the microscopic field $\bar{e}$ has meaning but which is small enough so as not to be considered macroscopically large. Van Vleck (31) refers to such a region as a "physically small" region and proves that in it the macroscopic field $\bar{E}$ is equal to the space-time average of the microscopic field $\bar{e}$. Therefore, one may treat this region as a continuum in so far as the macroscopic observables electric field and polarization are concerned.

Each dipole in such a region will be acted on by an average force given by

$$\bar{f} = \bar{p} \cdot \bar{\nabla} \bar{E} \tag{82}$$

where $\bar{p}$ is the dipole moment of the dipole. The force per unit volume $\bar{F}'$ averaged over this region is therefore given by

$$\bar{F}' = \bar{P} \cdot \bar{\nabla} \bar{E} \tag{83}$$

where $\bar{P}$ is the average dipole moment per unit volume or commonly the polarization. The force $\bar{F}'$ of equation (83) is called the volume force or the ponderomotive force due to Kelvin.

Equation (83) may be written in a more restricted form. In the case where $\bar{P}$ is directly proportional to $\bar{E}$, the following substitution may be made for $\bar{P}$:

$$\bar{P} = \epsilon_0 (K-1) \bar{E} \tag{84}$$

The result of this substitution is

$$\bar{F}' = \epsilon_0 (K-1) \bar{E} \cdot \bar{\nabla} \bar{E} \quad . \tag{85}$$

The vector identity,

$$(\bar{\nabla} \times \bar{E}) \times \bar{E} \equiv \bar{E} \cdot \bar{\nabla} \bar{E} - \frac{1}{2} \bar{\nabla} E^2 \equiv 0 \tag{86}$$

valid for any electrostatic case, may be employed to give

$$\bar{F}' = \frac{\epsilon_0}{2} (K-1) \bar{\nabla} E^2 \quad . \tag{87}$$